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Design and Performance of a Field Exposure System for Evaluation of the Ecological Effects of SO2 on Native Grassland

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16. ABSTRACT

We describe the design and performance of two replicate field exposure systems for evaluating the ecological effects of chronic SO_2 exposure on 0.5 ha plots of native Montana grassland. The SO_2 was supplied at a constant rate to each plot through a network of 2.5-cm inside diameter aluminum pipes set parallel to and approximately 0.7 m above the ground with 0.8-mm SO_2 release holes at 3-m intervals. The resulting SO_2 concentrations on the plots varied with micrometeorological conditions. Fumigations were essentially continuous from April through October for 5 yr on one set of four plots and for 4 yr on another set of four plots. Frequency distributions of SO_2 concentrations were approximately log-normal and qualitatively similar to those from various area and point sources. The diurnal pattern of SO_2 concentrations was strongly correlated with the reciprocal of windspeed. Nighttime concentrations averaged almost double daytime concentrations. The diurnal concentration pattern resembled an area source more closely than a point source, a direct result of using constant SO_2 source strength. Plume strikes could be simulated by turning the system on only during pre-specified meteorological conditions. Similarly seasonal variations in area source concentrations could be simulated by varying source strength on the plots. The data from 5 years use of the system indicate that the fine-scale behavior of SO_2 concentrations would be realistic.

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DESIGN AND PERFORMANCE OF A FIELD EXPOSURE SYSTEM FOR EVALUATION OF THE ECOLOGICAL EFFECTS OF SO_2 ON NATIVE GRASSLAND

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Abstract. We describe the design and performance of two replicate field exposure systems for evaluating the ecological effects of chronic SO_2 exposure on 0.5 ha plots of native Montana grassland. The SO_2 was supplied at a constant rate to each plot through a network of 2.5-cm inside diameter aluminum pipes set parallel to and approximately 0.7 m above the ground with 0.8-mm SO_2 release holes at 3-m intervals. The resulting SO_2 concentrations on the plots varied with micrometeorological conditions. Fumigations were essentially continuous from April through October for 5 yr on one set of four plots and for 4 yr on another set of four plots. Frequency distributions of SO_2 concentrations were approximately log-normal and qualitatively similar to those from various area and point sources. The diurnal pattern of SO_2 concentrations was strongly correlated with the reciprocal of windspeed. Nighttime concentrations averaged almost double daytime concentrations. The diurnal concentration pattern resembled an area source more closely than a point source, a direct result of using constant SO_2 source strength. Plume strikes could be simulated by turning the system on only during pre-specified meteorological conditions. Similarly, seasonal variations in area source concentrations could be simulated by varying source strength on the plots. The data from 5 years use of the system indicate that the fine-scale behavior of SO_2 concentrations would be realistic.

1. Introduction

For several years the U.S. Environmental Protection Agency has investigated the long-term consequences of chronic sulfur dioxide exposure to native grasslands of the northern Great Plains. The ecological field experiments in this program were the first to expose large areas of a natural ecosystem to controlled levels of a gaseous pollutant. They were designed to determine the effects of SO_2 on biomass dynamics (plant, arthropod, and small mammals), nutrient cycling, plant and animal community structure, insect and fungal diseases of plants, pollination systems, lichens, and upon several physiological and biochemical functions.

Field exposure systems (Zonal Air Pollution Systems, ZAPS) were designed to permit continuous SO_2 fumigation of a minimally disturbed grassland plot during an entire growing season. Versions of ZAPS have been successfully adapted to other situations including assessment of effects on crops (Miller *et al.*, 1979) and on grasslands in the Netherlands (Bovenkerk *et al.*, 1979). A second generation ZAPS will find major application in the National Crop Loss Assessment Network (NCLAN) (Lewis and Shriner, 1980). In this paper, we describe the design and performance of the experimental system and characterize the exposure history of the grassland plots during the 5-yr course of the field studies.

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2. Materials and Methods

Experiments were conducted within two 10-ha grassland exclosures in the Custer National Forest in southeastern Montana located in Sec. 9, T7S, R47E (45° N, 106° W). Livestock were excluded to protect them from injury and to prevent equipment damage. Sulfur dioxide fumigation of plots on the first exclosure (ZAPS I) was initiated in May 1975. Fumigation of the second set (ZAPS II) was started in April 1976.

Plots were usually fumigated from early April through October. Sulfur dioxide treatments began when the 10-day running average of air temperature exceeded 5.0 °C and terminated when the 10-day running average fell to 5.0 °C. At 5 °C, photosynthetic activity of the dominant plants on the sites (Western Wheatgrass (*Agropyron smithii*), Prairie Junegrass (*Koeleria cristata*), and Sandberg Bluegrass (*Poa secunda*)) is generally much less than at higher, typical season temperatures (Lee *et al.*, 1975; Singh *et al.*, 1980). The 10-day moving average ensured that unseasonal weather was not a factor in the fumigation schedule (Lee and Lewis, 1978). The experimental fumigation lasted for five growing seasons on ZAPS I and for four seasons on ZAPS II.

2.1. The Gas Delivery System

Each gas delivery system consisted of a network of 2.5-cm aluminum pipes (schedule 40, alloy 6061-T6) set parallel to and approximately 0.7 m above the ground and supported at 6.1-m intervals by pipes driven 0.8 m into the ground (Figure 1). Sulfur dioxide release points (0.8 mm holes which released the gas horizontally) were placed at 3-m intervals

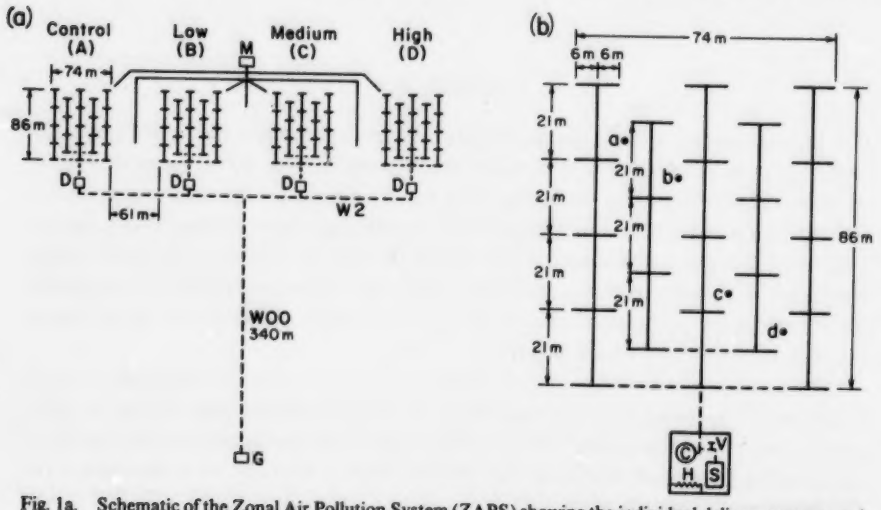

Fig. 1a. Schematic of the Zonal Air Pollution System (ZAPS) showing the individual delivery systems and the common monitoring and electrical systems. CONTROL, LOW, MEDIUM, and HIGH indicate relative SO_2 exposures on the experimental plots. (b) Expanded view of a single plot.

Key: (*a, b, c, d*) real-time monitoring locations; (C) compressor; (D) delivery stations; (G) 6 kw diesel generator; (H) 1 kw heater; (M) central monitoring station; (S) sulfur dioxide tanks; (V) valve; (WOO) three conductor o/o copper wire; (W2) three conductor 2 aluminum wire.

so that no location within a plot was more than 5.5 m from a source. A continuous flow of air through the lines was maintained by a helical compressor (Becker model SV 80-1*). Gas delivery equipment was housed in a heated shed with the temperature kept high enough so that the vapor pressure of the SO_2 in the tanks was always above the desired release pressure. Pressure regulators on the tanks and a flow controller (Brooks model 8944) and flow meter (Brooks 1110) at the injection point assured the desired SO_2 flow rate. Air pressure in the lines varied less than 5% across the system. The only ground level obstructions within the plots were the pipe network supports.

Each ZAPS study site consisted of four 0.5-ha treatment plots located along a line with 61-m intervening buffer zones to reduce interference between plots. Since the plots were relatively close their ecological features were similar. On all but one plot, SO_2 was bled into the air stream of the pipe network. The fourth plot (control) received no SO_2 except for occasional drift from other plots.

To achieve desired ambient concentrations, treatment plots received SO_2 at constant rates of 42.5, 99.1, and 198.2 standard liters per hour per plot. By convention, these treatments were identified as LOW (B), MEDIUM (C), and HIGH (D); the CONTROL was designated (A). Sulfur dioxide concentration in the delivery lines was approximately 0.5–1% (volume) on the HIGH plots and proportionately less for plots with lower SO_2 flow rates. Concentrations in the lines determined average atmospheric concentrations on each plot. Actual atmospheric concentrations were allowed to vary with meteorological conditions.

2.2. Real-time monitoring system

Sulfur dioxide concentrations were monitored at various plot locations (Figure 1) throughout the growing season. While not all locations on all plots were monitored, location *c* was continually monitored on all plots to permit interplot comparisons. Use of multiple locations within some plots allowed intraplot comparisons. Air samplers were placed 30–35 cm above the ground (approximate canopy height).

Real-time sulfur dioxide concentrations at 1, 15, and 31 cm above the ground were measured at a common central point on all plots during the 1978 growing season (Wong *et al.*, 1980, Table I).

Sulfur dioxide concentrations on the plots were continuously monitored by recording the output from a Meloy model SA160-2 flame photometric (FDP) sulfur gas analyzer operating in logarithmic mode. Ambient air samples were continuously drawn through eight teflon sample lines by a time-share device (Adgo Co.). One line passed through a zero-SO_2 filter, providing a check for drift. The other seven lines extended to monitoring locations on the plots (at least one per plot).

At the time-sharing device, the ambient air streams were diverted either to the sulfur gas analyzer or to an external exhaust. Each sample line was monitored continuously

* Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

TABLE I

Measurement periods of vertical profiles in SO_2 concentration on ZAPS (Zonal Air Pollution System)

ZAPS	Treatment plots		
	High treatment	Medium treatment	Low treatment
I	7/26/78 → 8/29/78 (34 days)	7/13/78 → 7/26/78 (14 days)	6/27/78 → 7/13/78 (17 days)
II	7/14/78 → 7/27/78 (14 days)	7/27/78 → 8/08/78 (13 days)	8/09/78 → 8/28/78 (19 days)

for 7.5 min of each hour. Lines were sampled sequentially, from lowest to highest concentration treatment plots.

The sulfur gas analyzer was calibrated against NBS-SRM 1627 permeation tubes using either an Analytical Instrument Development, Inc., Model 303 gas mixing system (before June 1977) or a Metronics Model 330 Dynacalibrator (June 1977 and later). Span checks were conducted every 2 weeks. Multipoint recalibrations were conducted if the span check detected an error of 5% or greater. The need for recalibration was uncommon.

The Dynacalibrator used ambient air as a carrier for the calibration gas. During the 1977 season, air was drawn from within the monitoring shed, where carbon dioxide concentrations approached 485 ppm (Lee *et al.*, 1979), considerably above the ambient concentration of 330 ppm. Since CO_2 can significantly depress response characteristics of FPD sulfur gas analyzers, SO_2 concentrations in 1977 were corrected using relationships determined by Weber *et al.*, 1980. Upper and lower bounds for the CO_2 interference were estimated by assuming that CO_2 levels were 485 ppm or 330 ppm during calibrations. The problem was solved in 1978 by attaching the Dynacalibrator pump inlets to tubing running to external air.

Calibration curves were constructed relating analyzer response to the logarithm of SO_2 concentration typically became nonlinear at SO_2 concentrations below roughly 45 $\mu g\ m^{-3}$. Available calibration equipment did not permit determination of the precise nature of the nonlinearity. Since linear extrapolation on these calibration curves to zero analyzer response led to estimated SO_2 concentrations equal to or greater than 20 $\mu g\ m^{-3}$, it is probable that linear extrapolation from calibration curves leads to an nearly all cases, was probably between 2 $\mu g\ m^{-3}$ (possible background concentrations) and the value estimated from the calibration curve. These values were used to specify upper and lower bounds for any SO_2 concentration falling in the questionable region of the calibration curves.

To ascertain the extent of sample dilution by permeation of SO_2 out of the lines or leaking of air into the lines during transit from the monitoring location to the time share in the monitoring building, a known concentration of SO_2 from a permeation type calibrator was sampled first directly at the analyzer and then after passing the full length (119 m) of one of the Teflon sampling lines normally used to conduct ambient air samples

to the analyzer. Any loss of SO_2 during transmission would be discernible by comparing the two concentrations detected by the sulfur analyzer. No significant degradation of the SO_2 sample was detected from transmission through the sample line.

2.3. SYNTHESIS PROCEDURES

The 7.5-min average analyzer output from each sampling location was used to estimate weekly, monthly and seasonal geometric means (GM), standard geometric deviations (SGD), and arithmetic means (AM). Since the SO_2 analyzer output was linear in the logarithm of the concentration, these estimates were based on the average logarithm and thus the median 7.5-min concentration.

To estimate 1-hr, and 3-hr peaks, continuous readings from each location were needed. A computer program was used to estimate these from 7.5-min median concentrations. The synthesis procedure consisted of several steps. First, the median value for each 7.5-min interval as read from the strip chart was entered into the computer. The zero-filter reading (usually zero) was subtracted to correct for drift and this value entered into the calibration equation.

All data were then scaled to the same geometric mean. After all data for a week were entered, the GMs for all sampling locations for that week were calculated. The concentration for each 7.5-min interval for an arbitrarily assigned standard location, s, was estimated by multiplying the measured concentration at location i by the ratio of weekly GMs.

$$C_s\,(\text{est}) = C_i\,(\text{meas})\frac{\text{GM}_s}{\text{GM}_i}. \tag{1}$$

This was equivalent to centering the normal distributions of logarithms at the same value. Analysis of ZAPS data indicated that SO_2 concentrations at various locations (except for the control plot) varied similarly when normalized to the same GM (Lee and Lewis, 1978).

The estimated 7.5-min medians at the standard locations were combined into averages for non-overlapping time intervals of 1 hr and longer. For example, for a given day, all 7.5-min medians were averaged for various interval (i.e., midnight to 1 a.m., 1 a.m. to 2 a.m., etc.). This standard procedure (Larsen, 1971) yields 24 1-hr averages and eight 3-hr averages per day. Averages for each nonstandard location, i, were estimated by multiplying standard averages by the ratio $\text{GM}_i : \text{GM}_s$.

Concentrations for the plot receiving no direct SO_2 input (CONTROL, plot A) were excluded from this averaging process since these were usually below the detection threshold of the analyzer. For this sampler, the 7.5-min median was used to represent the entire hour and 3-hr averages were based on these. Thus, averages for this sampler were much more sensitive to short term fluctuations, and consequently less reliable than averages for other samplers.

Errors in SO_2 concentration estimates caused by CO_2 interference and nonlinearity of calibration curves could not be expressed as simple percentages of estimated concentrations. Therefore, each strip chart value was entered into two calibration equations.

In the HIGH run, the caibration equation was adjusted to yield a maximum estimate likely to result from the quantifiable sources of uncertainty. The calibration equation for the LOW run was adjusted to yield the corresponding minimum estimate. Summary statistics were computed for both the HIGH and LOW runs. We assume that the range of values presented brackets the true values.

2.4 Time integrated system to assess spatial variation

In order to determine the horizontal distribution of SO_2, a series of Huey sulfation plates were set out at various locations at canopy height (35 cm) over the entire study site. The plates, which absorb ambient sulfur dioxide and retain it as lead sulfate, were placed on poles for a specified length of time (as indicated below), collected, and then analyzed for sulfate content. Results of this analysis were used to describe the SO_2 distribution pattern over the ZAPS site.

This study was composed of INTENSIVE and EXTENSIVE portions. The primary purpose of the EXTENSIVE study was to index the general relative concentrations of SO_2 at various points in horizontal space on the plots. The treatment plots and buffer zones were investigated as separate plots, making a total of seven per ZAPS site. Plates were widely spaced over each of the seven plots (Figure 2a). One pole was placed in the center of each plot quadrant. A fifth pole was set next to the real-time sulfur analyzer sample position *c* (Figure 1) on plots A, B, C, and D, and in the geometric center of the

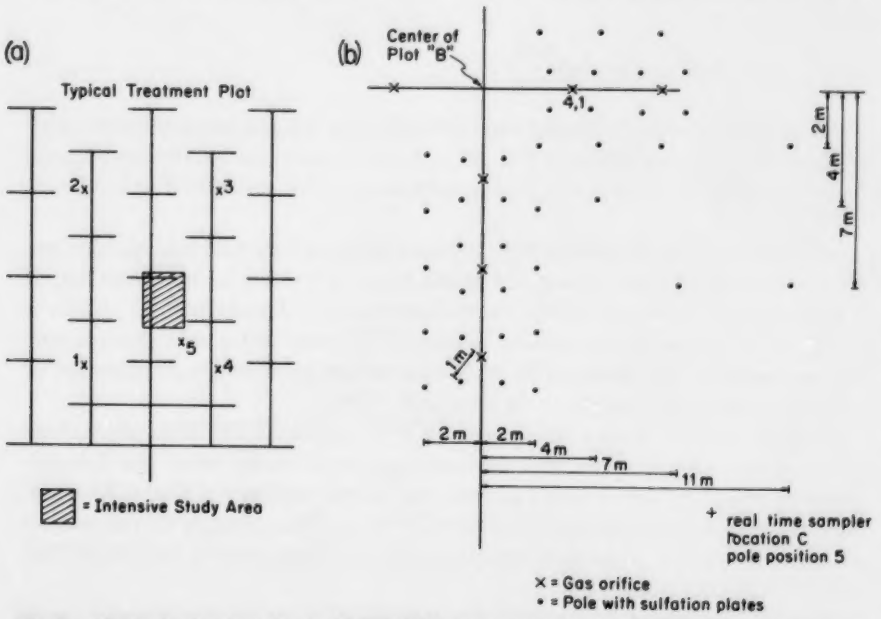

Fig. 2a. Monitoring locations for the EXTENSIVE study and location of INTENSIVE study area on ZAPS, plot B. (b) Spatial relationship of gas delivery orifices and sulfation plate monitoring locations in the INTENSIVE study.

interval plots. The study was repeated on both ZAPS I and II for four periods in 1977 (May 24 to June 24, June 25 to July 25, July 26 to August 25, August 26 to September 25).

The major purpose of the INTENSIVE part of the study was to determine the disperson pattern of the SO_2 immediately after leaving the delivery system. Sulfation plates were placed in a tight pattern 35 cm above the ground around five gas delivery orifices on plot B (Figure 2b). The study was repreated for both ZAPS I and II for three test periods in 1977 (July 5 to August 5, August 6 to September 5, September 5 to October 6).

In all instances, the sulfation plates were held parallel to and 35 cm above the ground with the open face down. Sulfate determination of the exposed plates was conducted by the Environmental Management Planning Laboratory in Nashville, Tennesee.

2.5. Micromet monitoring system

Air temperature and relative humidity were measured with a hydrothermograph (Bendix Model 594). Wind speed and direction were monitored at 2.1 m above ground with a DC-powered portable system consisting of a balsa vane and a three-cup anemometer (Ecowind IIIB, Wong Laboratories). The threshold for both indicators was 33.5 cm s^{-1}. Wind direction was recorded as an analog signal on a scale marked N-E-S-W-N, while wind speed was recorded as an integrated signal, a mark for every 0.8 km of air passage. Data were recorded on a Gulton recorder (Model 288/F2146). All instrumentation was placed in an instrument shelter 1.5 m above ground.

3. Results and Discussion

3.1. Anticipitated plume behavior

By utilizing many small, elevated, dilute point sources (over 250 per 0.5 ha), adequate dilution of SO_2 at ground level was ensured and, in effect, an area source was created above the plots. This prevented step-function changes in concentrations in time and space ('hot spots'), except at short distances from a source.

Theoretical insight into the behavior of a plume near a single release point has been obtained by calculating the densimetric Froude number (Albertson *et al.*, 1950), defined by

$$F = \frac{U}{\sqrt{\frac{\Delta P}{P} Dg}} \,. \tag{2}$$

The required parameters are the discharge velocity U, the jet diameter D, the ratio of excess mass density ΔP to the ambient density P, and the gravitational acceleration constant g. This dimensionless number represents the ratio of inertial to buoyant forces. For values of F greater than 40, the plume began to approach neutral buoyancy; i.e., the

SO_2 could be considered a trace constituent whose destiny is determined by the fate of its ambient air parcel. For the ZAPS plot with the highest SO_2 concentration (Plot D), F was larger than 10000. Thus, the SO_2 concentration was controlled by wind and turbulence and did not settle to earth and accumulate.

3.2. Frequency distribution curves

Concentration frequency distributions for ZAPS II, 1978 were compared with two major area sources (Chicago and San Francisco), point sources (660 megawatt coal-fired generating plants at Colstrip, MT) and the smelters at Sudbury, Ontario in Figure 3. All curves were approximately log-normal and qualitatively similar. Concentrations for the

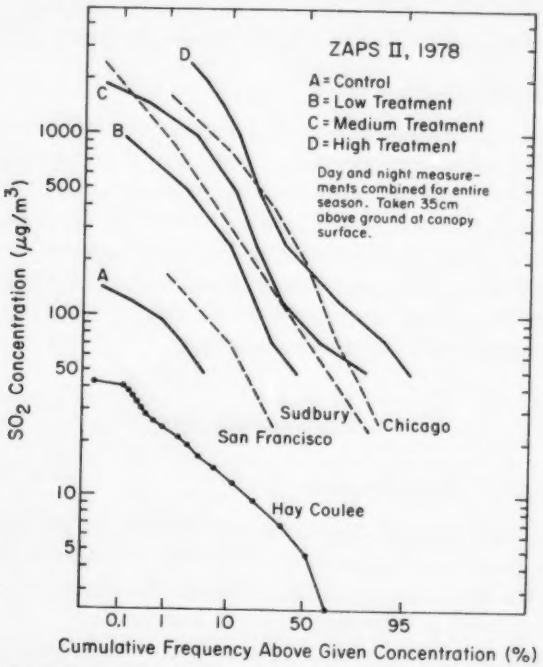

Fig. 3. Frequency distributions of 7.5-min SO_2 concentrations monitored on ZAPS II, 1978 (HIGH run) compared with those monitored in Chicago, San Francisco, (1962–67) (Health, Education, and Welfare, 1970), and Hay Coulee (12 km S.E. of Colstrip power plants, 1977–79), and Sudbury, 1964–1968. Hay Coulee data are from Ludwick *et al.*, 1980; Sudbury data are from Dreisinger and McGovern (1970).

area and point sources were measured higher above the ground than those on ZAPS so direct quantitative comparisons are inappropriate. The curves are typical of pollutant frequency distributions (Health, Education and Welfare, 1970; Larsen, 1969). Such distributions are characterized by the geometric mean (GM) and standard geometric deviation (SGD). These parameters summarize the temporal frequency of occurrence of concentrations, and thus represent a measure of dose. Typical values for actual pollutant distributions and for ZAPS are given in Table II.

TABLE II

Geometric mean SO_2 concentrations (GM, $\mu g\ m^{-3}$) and their Standard Geometric Deviations (SGD) for several urban areas and ZAPS. ZAPS data are based on 7.5-min medians 1975–1979 (ZAPS I) or 1976–1979 (ZAPS II). Urban areas based on 5-min averages, 1962–1967 (Health, Education, and Welfare, 1970)

Site	GM	SGD	1-hr peak	24-hr peak
Chicago	280	2.2	4530	2115
ZAPS II D	185–195	2.6–3.0	4410–5845	1155–1410
ZAPS I D	170–180	2.7–2.9	10650–10740	1384–1710
Philadelphia	150	2.4	2760	1235
ZAPS II C	105–110	2.3–2.5	2840–3695	645–740
ZAPS I C	105–110	2.8	3950–3975	740–830
Washington	105	2.2	1660	670
St. Louis	75	2.8	2575	695
ZAPS II B	60–65	2.0–2.4	1525–1915	345–370
ZAPS I B	55–60	2.4–2.7	2540	440–485
Cincinnati	45	2.9	1530	480
Los Angeles	35	2.3	775	270
Denver	35	2.1	965	160
ZAPS II A	10–30	1.3–3.3	760–780	70–80
ZAPS I A	15–25	1.5–2.8	600–645	115
San Francisco	15	2.9	695	215

In models of pollutant dispersion, source strength appears as a normalization constant, with the patterns of distribution determined by meteorological conditions (Turner, 1969). The resultant low variability of SGD over a range of GMs is evident for both the urban areas and ZAPS (Table II). The only dissimilar SGD is for the A plots which did not receive direct SO_2. The similarity of the SGDs indicates that the fine-scale variability of SO_2 concentrations throughout the study plots was realistic and reproducible.

3.3. Interseasonal Variation

Summaries of concentrations measured at canopy height on the ZAPS plots over the 5-yr experiment are given in Table III. Although SO_2 release rates were similar for all years, seasonal differences in weather caused year-to-year differences in SO_2 concentrations on the plots. Such differences also occur under ambient conditions.

3.4. Intraseasonal Variation

Sulfur dioxide concentrations varied considerably during the course of each growing season (Figure 4). The variability tended to increase with exposure concentration. The HIGH plots were particularly variable (Lee *et al.*, 1979; Preston *et al.*, 1980).

3.5. Diurnal Variation

Sulfur dioxide concentrations on the plots were higher and more variable during night hours than during daylight (Table IV, Figure 4) (Lee and Lewis, 1978; Lee *et al.*, 1979; Preston *et al.*, 1980). This was caused by generally poor mixing and low wind speeds at

TABLE III

Seasonal average SO_2 concentrations measured in the ZAPS plots at location *c* from 1975–1979 (low run-high run, $\mu g\ m^{-3}$). Single values are presented when low run and high run values rounded to the same value

Year	Measure	ZAPS I				ZAPS II			
		Control	Low	Medium	High	Control	Low	Medium	High
1975	G.M.[a]	25[f]	50[f]	90[f]	155[f]				
	S.G.D.[b]	1.5	2.4	2.7	2.7				
	A.M.[c]	25	75	145	255				
	1-hr[d]	255	740	1245	2055				
	3-hr[e]	255	460	1085	1595				
1976	G.M.	5– 20	40– 50	85– 90	150– 155	5– 30	60– 65	95– 100	150– 160
	S.G.D.	3.0, 1.5	3.6, 2.5	3.1, 2.7	2.9, 2.5	3.4, 1.2	2.3, 1.8	2.3, 1.9	3.0, 2.3
	A.M.	10– 25	90	170	265	10– 30	85	125	245
	1-hr	300	1410–1455	2935–3740	6100– 8155	140	1570–1710	2540–2890	4595–5195
	3-hr	175–180	855– 925	1895–2285	3950– 4990	90	925– 995	1550–1665	2700–3005
1977	G. M.	5– 35	75– 80	120– 130	190– 220	10– 35	65– 70	110– 115	175– 190
	S.G.D.	4.5, 1.7	2.2, 2.1	2.7	3.1, 2.5	4.2, 1.2	2.1, 1.9	2.2, 2.1	2.7, 2.4
	A.M.	25– 50	105– 110	220– 235	325– 355	25– 35	95– 100	155– 175	275– 300
	1-hr	600–645	1180–1295	2265–2355	3165– 3720	760–880	1525–1915	2840–3695	4410–5845
	3-hr	460–485	800– 945	1480–1640	2220– 2680	275–325	880–1040	1895–2285	3465–4295
1978	G.M.	25	65	120	200– 205	20– 30	60	105	210– 215
	S.G.D.	1.7, 2.1	2.7	3.1	3.2, 3.4	1.3, 2.3	2.3	2.6	2.8, 2.9
	A.M.	30	125	270	445	25– 30	90	185	410
	1-hr	600	2540	3950–3975	10650–10740	140	695	1315	2890
	3-hr	255	1615	2520	6790– 6815	90	510	1085	2055
1979	G. M.	20– 30	55– 60	115	170	20– 30	55– 60	115– 120	220– 225
	S.G.D.	0.8, 1.6	2.4, 2.1	2.5	2.4	2.9, 1.3	2.6, 2.1	2.7, 2.5	3.0, 2.8
	A.M.	35– 40	85	195	1030	30– 35	85– 90	195– 200	420
	1-hr	440	1050–1095	2020–2025	3190– 3200	450	920	1400	3530
	3-hr	305	635– 640	1215–1220	2030– 2060	240	595	1095	2625

[a] Geometric mean.
[b] Standard geometric deviation.
[c] Arithmetic mean.
[d] Highest 1-hr average concentration observed during the growing season.
[e] Highest 3-hr average concentration observed during the growing season.
[f] High run only for 1975.

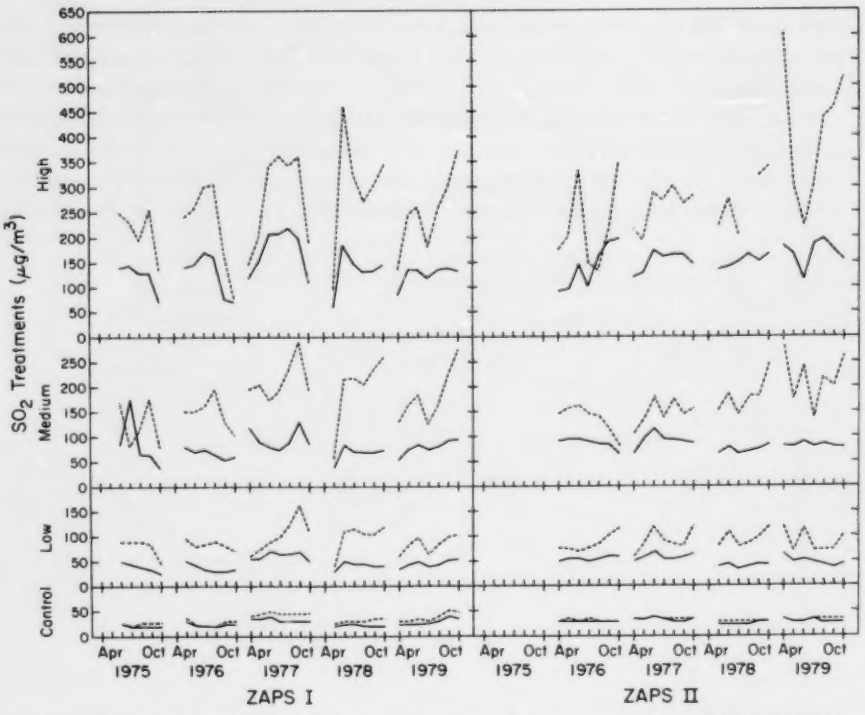

Fig. 4. Daytime and nighttime monthly geometric mean SO_2 concentrations on ZAPS (High Run). —— Daytime, ----- Nighttime.

TABLE IV

Day and night geometric mean SO_2 concentrations ($\mu g\ m^{-3}$, high run) on the ZAPS sites, 1975–1979 (based on 7.5-min median concentrations)

Site	Plot	Year											
		1975		1976		1977		1978		1979		Overall	
		Day	Night	Day	Night	Day	Night	Day	Night	Day	Night	Day	Night
I	A	20	25	20	25	35	45	25	30	30	40	25	30
	B	40	75	35	85	60	105	45	110	45	90	45	90
	C	75	115	60	145	90	215	70	220	80	180	75	170
	D	120	210	125	195	180	280	145	325	130	255	140	250
II	A			30	30	35	35	25	30	30	35	30	30
	B			55	90	60	90	40	95	50	85	50	90
	C			90	125	95	150	75	180	85	200	85	160
	D			140	205	155	255	150	295	160	365	150	275

night compared to more complete mixing and higher wind speeds during the day. The diurnal pattern of SO_2 concentrations was strongly correlated with the reciprocal of wind speed (Lee *et al.*, 1979; Preston *et al.*, 1980), as expected from dispersion models (Turner, 1969). Solar radiation accounted for a smaller amount of SO_2 variability, while temperature gradient and relative humidity were unimportant (Lee *et al.*, 1979). Average hourly sulfur dioxide concentration for each plot at 1, 15, and 31 cm above ground are shown in Figure 5. In general, sulfur dioxide concentrations were highest near the top

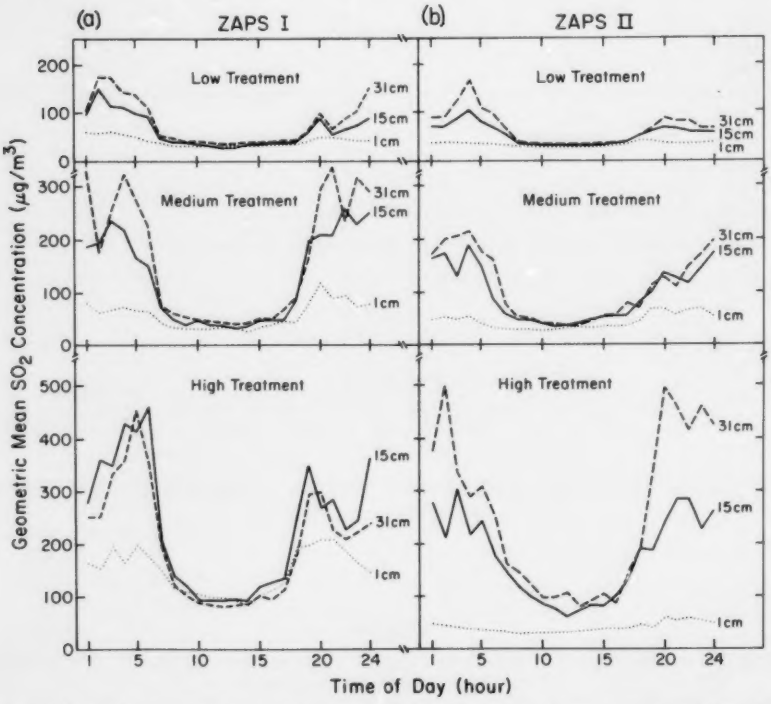

Fig. 5. Hourly SO_2 concentrations within the vegetation canopy during measurement periods specified in Table I.

of the plant canopy and decreased with decreasing height. Significant vertical stratification of sulfur dioxide occurred primarily at night when concentrations reached maximum levels. During daytime, concentrations were lower and vertical stratification was slight.

Canopy air flows can largely determine the rate of SO_2 transport to biota from the delivery system. Horizontal winds flowing over the canopy are slowed by frictional drag on the vegetation. Immediately above the canopy, the mean horizontal wind velocity decreases logarithmically with decreasing height. Within the canopy, a canopy-eddy layer exists in which wind speed decreases exponentially. In the lowest part of the plant-air layer, a logarithmic wind profile resumes with wind speed decreasing to zero (Inoue,

1963). The degree of vertical mixing of SO_2 at various vertical distances from the SO_2 delivery pipes on the ZAPS plots should vary directly with wind speeds at those heights and therefore should decrease as wind speeds decrease with decreasing height above ground level.

Condition of the vegetation surface may also influence the vertical SO_2 uptake rates. Because of higher relative humidity and dew formation at night, greater SO_2 uptake rates may occur even though stomatal resistance is higher at night. Greater uptake rates at night should enhance the vertical concentration gradient. Thus the observed diurnal variation in the vertical profile in SO_2 concentration can be explained by meteorological factors alone.

Diurnal fluctuations in SO_2 concentrations generally followed one of four patterns, with concentration peaks occurring at the following times:

(1) A single concentration peak between 1800 and 2400 (i.e., 6 p.m. and midnight),

(2) A single concentration peak between 2400 and 600,

(3) Two concentrations peaks, one between 1800 and 2400 and one between 2400 and 600,

(4) Low variable concentrations with no dominant peaks (rare).

Occurrence of the first three patterns was strongly correlated with patterns of wind speed. The fourth pattern usually occurred during days having high average constant wind speed.

3.6. Extensive sulfation plate study

To characterize the distribution of SO_2 concentrations in horizontal space on the ZAPS plots, we compared sulfation at the five pole positions with one way ANOVA (Preston and Gullett, 1979). Plot locations were considered treatments and test periods were considered replications.

On treatment plots, there were statistically significant differences in sulfation rates among pole positions only on the D plots. Here, pole 1 had higher sulfation rates than other poles on ZAPS I ($p < 0.05$), and poles 1 and 4 had higher sulfation rates than others on ZAPS II ($p < 0.05$).

The two factors likely to have had the greatest influence on the spatial distribution of SO_2 concentrations were the gas delivery system and wind. The gas delivery system was intended to deliver a spatially uniform concentration of SO_2 at canopy height. However, any drop in pressure along the delivery pipes would lower the concentrations of SO_2. Frictional forces would increase the chances of significant pressure drop with increasing distance from the pump. Such a pressure drop could cause the observed differences on the D plots. However, measurements on ZAPS I in May, 1975 indicated a pressure drop of less than 5%.

Similar results could also be produced by wind patterns. If wind displacement of SO_2 were equal (on the average) in all directions, positions at the periphery of fumigated plots would tend to get diluted while interior positions receiving drift from the periphery, would tend to have higher SO_2 concentrations. If wind displacement were not equal in all directions, peripheral points receving the greatest dilution would have the lowest SO_2

concentrations while those points receiving the greatest drift should have the highest averages. In addition, all plots may have their SO_2 supplemented by drift from other fumigated plots. The magnitude of drift can be gauged somewhat by sulfation measured on the A plots since these receive SO_2 by drift alone. The seasonal average concentrations on the A plots were about one-fourth of those on adjacent B plots (ZAPS I = 25.9%, ZAPS II = 27.2%). Thus, observed trends in sulfation may be explained by wind patterns (Preston and Gullett, 1979), but deficiencies in the gas delivery system cannot be ruled out.

3.7. Intensive sulfation plate study

Horizontal distribution of SO_2 was examined in finer detail in the INTENSIVE study. Positions were monitored 1, 2, 4, and 7 m from the gas delivery pipes.

If the crossed fumigation pipes are seen as dividing the study area into four sections, (Figure 2) then the poles 1- and 2-m from the pipes were sampled in three of the quadrants while the poles 4- and 7-m distant were sampled just in one. To assure data comparability, only the 1- and 2-m sulfation plates in the southeasterly quadrant were used to compare sulfation values at different horizontal distances from the delivery system.

Sulfation rates at increasing distances from SO_2 release points on ZAPS I and ZAPS II are compared in Table V. ZAPS II apparently had higher SO_2 concentrations

TABLE V

Average sulfation values and (standard error) at canopy height on low (B) treatment plots for plates, 1, 2, 4, and 7 m distant from delivery pipes. (All test periods combined, expressed as micrograms sulfate test period)

1m	2 m	4 m	7 m
ZAPS I			
230.28 (10.33)	220.24 (11.84)	236.33 (22.76)	228.33 (21.26)
ZAPS II			
321.40 (18.16)	273.38 (15.86)	277.67 (16.50)	258.00 (16.07)

within 1 m of the gas release points than at other locations on the plots. The horizontal dispersion of SO_2 appears to be quite uniform on both ZAPS sites beyond 1 m from the SO_2 release points.

3.8. Comparison of ZAPS SO_2 fumigation experimental sites to actual SO_2 sources

Figure 3 and Table II indicate that ZAPS simulates the geometric means, standard geometric deviations, and peaks of SO_2 concentrations from real sources. However, the temporal pattern of exposure can be quite different. For example, the diurnal pattern of the frequency of elevated SO_2 concentrations measured at Hay Coulee, 7.5 miles southeast of Colstrip, was unimodal with plume strikes occurring most often near midday and gradually decreasing in frequency to very low levels at night (Preston *et al.*, 1980).

This is the inverse of the diurnal pattern of concentrations observed on the ZAPS plots (Figure 5). At Hay Coulee, high SO_2 concentration was dependent upon wind transport from the stacks at Colstrip. Plume strikes tended to occur during the day when wind speeds were high and turbulent mixing brought the plume to the ground. On ZAPS, on the other hand, elevated wind speeds diluted SO_2 concentrations during the day. Though the ZAPS exposure regime appears to simulate the shape of the concentration frequency distribution curve at Hay Coulee reasonably well, it did not replicate the diurnal cycle of this point source.

For area sources, such as urban centers, the diurnal SO_2 concentration patterns are primarily determined by variations in mixing, wind speed and source strength (Holzworth, 1973; Munn and Katz, 1959). Concentrations in the vicinity of a point source are, in addition to these factors, highly dependent upon wind direction. Plume strikes are likely to be more frequent during the daytime because conditions often favor mixing while at night stable conditions tend to prevent mixing of the plume to the ground (Smith *et al.*, 1979). The most probable concentration near a point source is zero. High SO_2 levels are rare and unpredictable.

Thus, the meteorological factors determining the temporal pattern of exposure from ZAPS, as used in these experiments, were more similar to that of an area source than a point source. This was a direct result of using a constant SO_2 flow rate on each exposure plot. The occurrence of plume strikes can be simulated by activating the system only during pre-specified meteorological conditions. Similarly, seasonal variations in area source strength can be simulated by varying the SO_2 flow rate. The data from 5 yr' use of the system indicate that, during such simulated episodes, the fine-scale behavior of SO_2 concentrations would be realistic. Similar approaches are being used for ZAPS-derived systems (Lewis and Shriner, 1980; Miller *et al.*, 1980). Thus, ZAPS is a valuable tool for evaluating plant response to realistic air pollutant exposures under minimally disturbed field conditions.

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